Annual Report of the International Nuclear Energy Research Initiative OSMOSE Project (FY05)

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Project Title: OSMOSE – An Experimental Program for Improving Neutronic Predictions of Advanced Nuclear Fuels

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Lead Collaborating Investigating Organization: CEA-Cadarache
Lead Collaborating Principal Investigator: Jean-Pascal Hudelot

Other Collaborating Organizations: CEA-Valrho

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NTD/SIM: H. Khalil

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Project Status Summary:

The goal of the OSMOSE program is to measure the reactivity effect of minor actinides in known neutron spectra of interest to the Generation-IV reactor program and other programs and to create a database of these results for use as an international benchmark for the minor actinides. The results are then compared to calculational models to verify and validate integral absorption cross-sections for the minor actinides.

The OSMOSE program includes all aspects of the experimental program – including the fabrication of fuel pellets and samples, the oscillation of the samples in the MINERVE reactor for the measurement of the reactivity effect, reactor physics modeling of the MINERVE reactor, and the data analysis and interpretation of the experimental results.

Significant accomplishments in FY05 include: **1**. The fabrication of eight of the OSMOSE samples at CEA-Valrho and shipment of the 8 samples to CEA-Cadarache. Six samples were shipped in July and two Np samples were shipped in September**. 2.** Calculations and experiments in support of characterization of the MINERVE reactor and for safety authorization were completed. Calibration of the pilot rod and measurements using the U-235 and Boron calibration samples were performed in the R1-UO2 configuration. **3.** An initial series of calculations of the reactivity-worth of the samples in the R1-UO2 core configuration were completed using the REBUS model. The calculations are based on the specifications for fabrication, so they are preliminary until sampling and analysis have been completed on the fabricated samples. The estimates indicate a range of reactivity effect from -22 pcm to +25 pcm compared to the natural U sample. **4. Oscillation measurements using the first 8 OSMOSE samples began in September in the R1-UO2 configuration.**

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1. INTRODUCTION

The design of nuclear systems has shifted over the years from a "test and build" approach to a much more analytical methodology based on the many advances in computational techniques and nuclear data. To a large extent current reactors can be calculated almost as well as they can be measured. This is due in particular to the high quality nuclear data available for the few major isotopes which dominate the neutronics of these systems. Nevertheless, most of the future nuclear systems concepts and advanced fuels development programs currently underway use significant quantities of minor actinides to address modern day issues such as proliferation resistance and low cost. For example, high burnup fuels contain large quantities of americium and curium. Systems designed for plutonium and minor actinide burning are very sensitive to uncertainties in americium and curium data. There are also several other programs where the minor actinide data are essential. These include the Accelerator Transmutation of Waste concepts, Generation-IV concepts, and Burnup Credit programs.

The need for better nuclear data have been stressed by various organizations throughout the world, and results of studies have been published which demonstrate that current data are inadequate for designing the projects under consideration [1] [2]. In particular, a Working Party of the OECD has been concerned with identifying these needs [3] and has produced a detailed High Priority Request List for Nuclear Data. The first step in obtaining better nuclear data consists of measuring accurate integral data and comparing it to integrated energy dependent data: this comparison provides a direct assessment of the effect of deficiencies in the differential data. Several US and international programs have indicated a strong desire to obtain accurate integral reaction rate data for improving the major and minor actinide cross sections. Specifically, these include: ^{232}Th, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm, and ^{247}Cm. Data on the major actinides (i.e. ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am) are reasonably well-known and available in the Evaluated Nuclear Data Files - (JEF, JENDL, ENDF-B). However, information on the minor actinides (i.e.^{232}Th, ^{233}U, ^{237}Np, ^{238}Pu, ^{242}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm, and ^{247}Cm) is less well-known and considered to be relatively poor in some cases, having to rely on models and extrapolation of few data points. This is mainly due to the difficulty of obtaining relatively pure samples of sufficient quantity (up to about one gram) to perform reliable reaction rate measurements.

A large and exhaustive experimental program has been planned in the MINERVE reactor facility at CEA-Cadarache. One of the programs – OSMOSE (Oscillation in Minerve of Isotopes in Eupraxic Spectra) – aims at obtaining in different experimental lattices a single and accurate experimental database for separated heavy nuclides.

The objective of the OSMOSE program is to measure very accurate integral reaction rates in representative spectra for the actinides important to future nuclear system designs and to provide the experimental data for improving the basic nuclear data files. These data will support advanced reactors designed for transmutation of waste or plutonium burning, sub-critical systems such as found in advanced accelerator applications, and waste disposal

and treatment programs in the area of criticality safety. The OSMOSE program is very generic, in the sense that it will measure these reaction rates over a broad range of isotopes and spectra and will be used to provide guidance to all nuclear data programs in the world. The data will provide information valuable to a large number of projects as noted above.

The OSMOSE program [4] will provide precise experimental data (integral absorption cross-sections) for a majority of the heavy nuclides important to reactor and nuclear fuel cycle physics - ^{232}Th, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm, and ^{245}Cm. Table 1 shows the isotopes of interest in the OSMOSE program and highlights which isotopes are critical for the various programs. Table 2 shows the target improvements in the quality of the nuclear data for the listed actinide isotopes. The study of these nuclides is performed on a large range of neutron spectra corresponding to specific experimental lattices (thermal, epithermal, moderated/fast, and fast spectra).

The OSMOSE experimental program will produce very accurate sample worth measurements for a series of actinides in various spectra, from over-moderated thermal spectra to fast spectra. The objective of the analytical program is to make use of this experimental data to establish deficiencies in the basic nuclear data libraries, identify their origins, and propose paths towards correcting them, in coordination with international nuclear data programs.

TABLE 1
OSMOSE Program – Isotopes of Interest

	JEFF3 validation	Criticality Burn-up credit	Pu recycling	Transmutation and incineration	Decay Heat power	Subsurface long-term Storage	Reactivity loss per cycle	Thorium cycle
^{232}Th	⊗							⊗
^{233}U	⊗							⊗
^{234}U	⊗	⊗					⊗	
^{235}U	⊗	⊗				⊗	⊗	
^{236}U	⊗	⊗					⊗	
^{238}U	⊗	⊗					⊗	
^{237}Np	⊗	⊗		⊗		⊗	⊗	
^{238}Pu	⊗	⊗	⊗	⊗	⊗	⊗	⊗	
^{239}Pu	⊗	⊗	⊗	⊗	⊗	⊗	⊗	
^{240}Pu	⊗	⊗	⊗	⊗	⊗	⊗	⊗	
^{241}Pu	⊗	⊗	⊗	⊗	⊗	⊗	⊗	
^{242}Pu	⊗	⊗	⊗	⊗	⊗	⊗	⊗	
^{241}Am	⊗	⊗	⊗	⊗	⊗	⊗	⊗	
^{243}Am	⊗	⊗	⊗	⊗	⊗	⊗	⊗	
^{244}Cm	⊗	⊗	⊗	⊗	⊗	⊗	⊗	
^{245}Cm	⊗	⊗		⊗	⊗	⊗	⊗	

Table 2
Target Improvements in Nuclear Data for the OSMOSE Program

Actinide	Parameter	Current Uncertainty (at 1σ)	Target Uncertainty (at 1 σ)
^{233}U	η_{therm} $\eta_{epitherm}$	± 2500 pcm ± 4000 pcm	± 1500 pcm ± 2500 pcm
^{234}U	I_r σ_i^{th}	± 10 % ± 2 %	± 3 % ± 1.5 %
^{236}U	I_r	± 5 %	± 3 %
^{237}Np	I_r σ_i^{th}	± 7 % ± 3 %	± 2 % ± 1.5 %
^{238}Pu	I_r σ_i^{th}	± 9 % ± 2 %	± 4 % ± 1.5 %
^{239}Pu	η_{therm} $\eta_{epitherm}$	± 3000 pcm ± 4000 pcm	± 2000 pcm ± 2000 pcm
^{240}Pu	I_r	± 3 %	± 1.5 %
^{242}Pu	I_r	± 4 %	± 2 %
^{241}Am	I_r σ_i^{th}	± 7 % ± 3 %	± 2 % ± 1.5 %
^{243}Am	I_r	± 5 %	± 3 %
^{244}Cm	I_r	± 5 %	± 3 %
^{245}Cm	η_{therm}	± 4000 pcm	± 1500 pcm
^{232}Th	I_r	± 4 %	± 2 %

Ir = resonance integral, σ_i^{th}= microscopic capture cross section, η = reproduction factor

The measurement program is utilizing the MINERVE reactor at CEA-Cadarache, which is a low-power uranium fueled pool reactor. The normal accuracy for small-worth samples in this reactor is on the order of 1% for relative reactivity-worth measurements and 2% for absolute reactivity-worth measurements. The total uncertainty in the OSMOSE samples is estimated to be about 3% including the uncertainty in the isotopic composition. Reactivity effects of less than 10 pcm (0.0001 or approximately 1.5 cents) will be measured and compared with calibrations to determine the differential reactivity-worth of the sample. Accuracies in small reactivity effects this low are only achieved through oscillation techniques.

Four different neutron spectra will be created in the MINERVE facility: over-moderated UO_2 (representative of a fuel processing plant or flooded storage cask), UO_2 matrix in water (representative of LWRs), mixed oxide fuel matrix (representative of cores containing MOX fuels), and epithermal spectra (representative of under-moderated reactors). The different spectra are achieved by changing the lattice within the MINERVE reactor.

The OSMOSE program began in 2001 with the preparation of samples. Reactor modifications began in 2002 and were completed in 2003. The measurement program at MINERVE began in 2003 with the qualification of the MINERVE reactor after modifications were complete.

DOE is collaborating with CEA on the OSMOSE program through this project within the Generation-IV Reactor Program and as part of the International Nuclear Energy Research Initiative. ANL is serving as the lead laboratory on the U.S. side and CEA-Cadarache is the lead laboratory on the French side. The INERI project is focused on supporting the measurements to be conducted at CEA-Cadarache (through experimental support for conducting the measurements, pre-analysis and planning, and post-measurement data analysis activities). The DOE/CEA collaboration on the OSMOSE program includes the supply of separated ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{243}Am from DOE, the participation of DOE in the conduct of the experiments, and the development and comparison of analytic tools and models of CEA and DOE based on Monte Carlo and deterministic methods.

The INERI project has been divided into 4 distinct tasks – reactor modeling, sample fabrication, experimental measurements, and data analysis. Within these high level tasks, there are numerous sub-tasks such as reactor modeling of different core configurations and calculations for different core parameters. A lead laboratory has been identified for each high-level task and other supporting laboratory efforts are also noted. The roles and responsibilities for the tasks associated with the OSMOSE project are shown in Table 3.

2. PROJECT ORGANIZATION:

Roles and Responsibilities for activities and tasks associated with the OSMOSE project are as defined in Table 3.

Table 3
Roles and responsibilities for each organization

Task Description	Lead	Support
Task 1: Reactor Modeling	ANL	CEA-Cadarache
Task 2: Sample Fabrication	CEA-Valrhô	CEA-Cadarache
Task 3: Experiments	CEA-Cadarache	ANL
Task 4: Data Analysis	CEA-Cadarache	ANL

3. REACTOR MODELING

Objectives:

The goal of the experimental measurements is to produce a database of reactivity-worth measurements in different neutron spectra for the separated heavy nuclides. This database can then be used as a benchmark for integral reactivity-worth measurements to verify and validate reactor analysis codes.

The analytic effort is being performed by ANL and CEA personnel using MCNP and separate suites of reactor analysis codes. In this manner, a cross comparison can be performed on the results to identify potential errors in the cross-section evaluations in the numerical methods and assumptions used within the codes. This will allow the improvement of these codes.

The pre-analysis reactor modeling effort provides detailed foreknowledge for planning the experimental measurements. It also allows detailed models of the different core configurations to be assembled which can be used to support the data analysis of the experimental results. This modeling effort also provides the opportunity to thoroughly check the data on the reactor configuration including the fuel and structural materials, composition, geometry, and operating conditions.

Technical Status:

Monte Carlo and deterministic models of the MINERVE facility in the R1-UO2 and R1-MOX configurations were developed to assess core and safety parameters. The deterministic model is also used to calculate the reactivity worth of oscillation samples in the central channel of the core. The models are based on the composition and geometry specifications listed in the Material Specification Report for the MINERVE reactor [5].

The deterministic model is based on the REBUS code system [8]. REBUS has been used to solve the diffusion equation in XYZ geometry with the finite difference method. The DIF3D module of the code system is used for this purpose. The self-shielded cross sections used in REBUS are provided by the one-dimensional-transport-code-system WIMS-ANL 5.07 [9]. The WIMS and REBUS models are fully described in [7].

The physical size of the REBUS and MCNP model are the same. Figure 1 and Figure 2 show radial and an axial views of the complete geometry for the REBUS model.

The reactor modeling effort has supported the renewal of the safety documentation for the MINERVE facility. A new safety calculation scheme for the MINERVE facility and its associated experimental programs was assembled. The scheme is based on modeling the whole reactor with a Monte Carlo code and deterministic codes and includes the following:

1. TRIPOLI-4 Monte Carlo model for the R1-UO2 and R1-MOX core configurations, for the entire 3D reactor description,
2. MCNP-4C2 and MCNPX models for the R1-UO2 and R1-MOX core configurations, based on the same material balance report of the reactor as for TRIPOLI-4 model for the entire 3D reactor description,
3. APOLLO2 deterministic model for the R1-UO2 and R1-MOX core configurations, only for the 2D experimental zone description, and
4. REBUS deterministic model for the R1-UO2 and R1-MOX core configurations, for the entire 3D reactor description.

The Monte Carlo calculation schemes were used for the determination of the following safety parameters: reactivity worth of the control rods, importance in fissions of the experimental zone versus the driver zone, reactivity excess of the core, effective fraction of delayed neutrons (β_{eff}). The deterministic models were used to calculate the reactivity worth of the oscillation samples that could not have been determined with enough accuracy by using Monte Carlo calculations (up to about 30 pcm only).

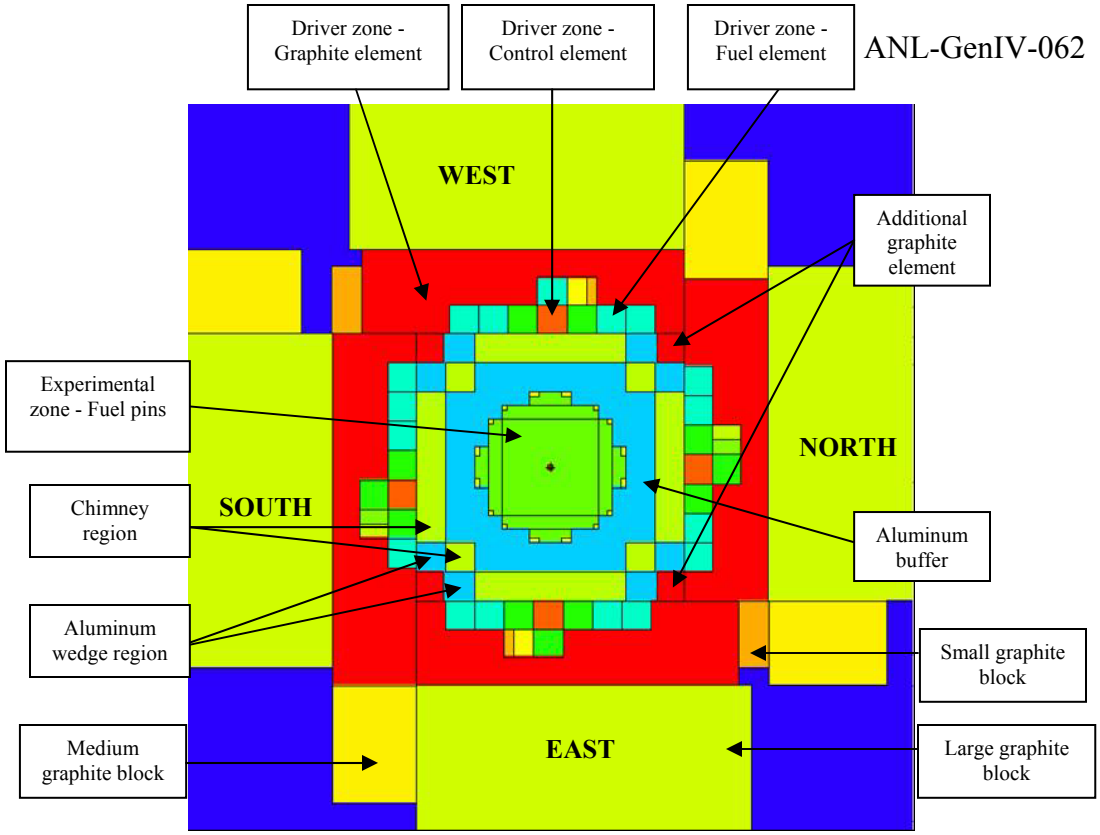

Figure 1: Radial view of the REBUS model in the R1-UO2 configuration

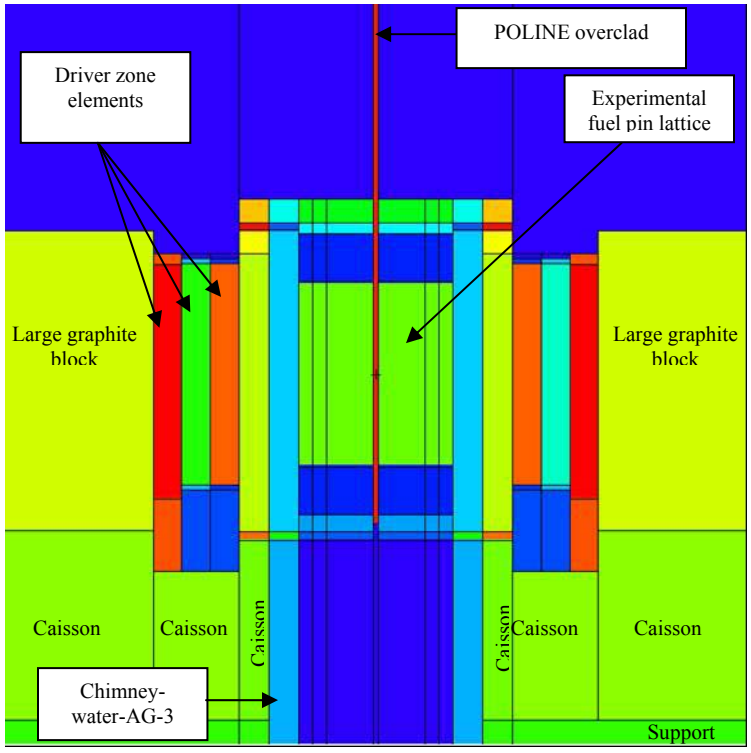

Figure 2: Axial view of the REBUS model in the R1-UO2 configuration

The safety report for the OSMOSE program in the R1-UO2 and R1-MOX core configurations was presented to the French safety authorities on April 28, 2005. The report did not include the two Curium samples that are planned to be oscillated in the OSMOSE program, but for which problems of shipment, transfer and radioprotection are still to be resolved. The two Curium samples will be addressed in a future report that will be presented to the safety authorities in 2006. In the end, the safety authorization for the start of the OSMOSE program was granted on June 21, 2005.

In addition to the safety authorization basis, the calculational scheme has been used to predict the reactivity effect of the OSMOSE samples in the R1-UO2 configuration. An initial series of calculations of the reactivity-worth of the OSMOSE samples in the MINERVE reactor with the R1-UO2 core configuration were completed. The reactor model was generated using the REBUS code developed at Argonne National Laboratory.

The results of the calculations are shown in Table 4 and graphically in Figures 3 and 4. The results for the reactivity-worth of the samples are compared to the natural uranium sample by subtracting the natural U sample value from the OSMOSE sample value. In this manner, the natural U sample shows a zero value for the reactivity-worth. The samples that show a positive reactivity-worth have a larger reactivity effect than natural uranium. Samples that have a negative reactivity-worth have a larger integrated absorption cross-section or a smaller fission cross-section than natural uranium (i.e. a larger value of Σ_a-$\upsilon\Sigma_f$).

The calculations are based on the specifications for fabrication, so they are considered preliminary until sampling and analysis have been completed on the fabricated samples. The estimates indicate a range of reactivity effect from -22 pcm to +25 pcm compared to the natural U sample. Results of the calculations are presented in [19].

Table 4:
Reactivity Worth of OSMOSE samples

Sample	Isotope Mass (g)	Keff	ρ (pcm)	Reactivity-worth (pcm)
Th-232 (1)	44.2	1.000528	52.8	-28.3
Th-232 (2)	2.1	1.000798	79.8	-1.3
U-233	0.5	1.000945	94.5	13.3
U-234	0.32	1.000788	78.8	-2.4
URE	0.4*	1.001001	100.0	18.9
U-nat	44.3	1.000812	81.2	0
Np-237 (1)	0.1	1.000786	78.6	-2.6
Np-237 (2)	0.6	1.000688	68.8	-12.3
Pu-238	0.4	1.000701	70.1	-11.0
Pu-239	0.6	1.000939	93.9	12.7
Pu-240	0.16	1.000723	72.3	-8.9
Pu-241	0.08	1.000840	84.0	2.9
Pu-242	0.5	1.000784	78.4	-2.7
Am-241 (1)	0.06	1.000755	75.4	-5.7
Am-241 (2)	0.2	1.000663	66.3	-14.8
Am-243	0.1	1.000789	78.9	-2.3

*This is the mass of U-236 in the sample.

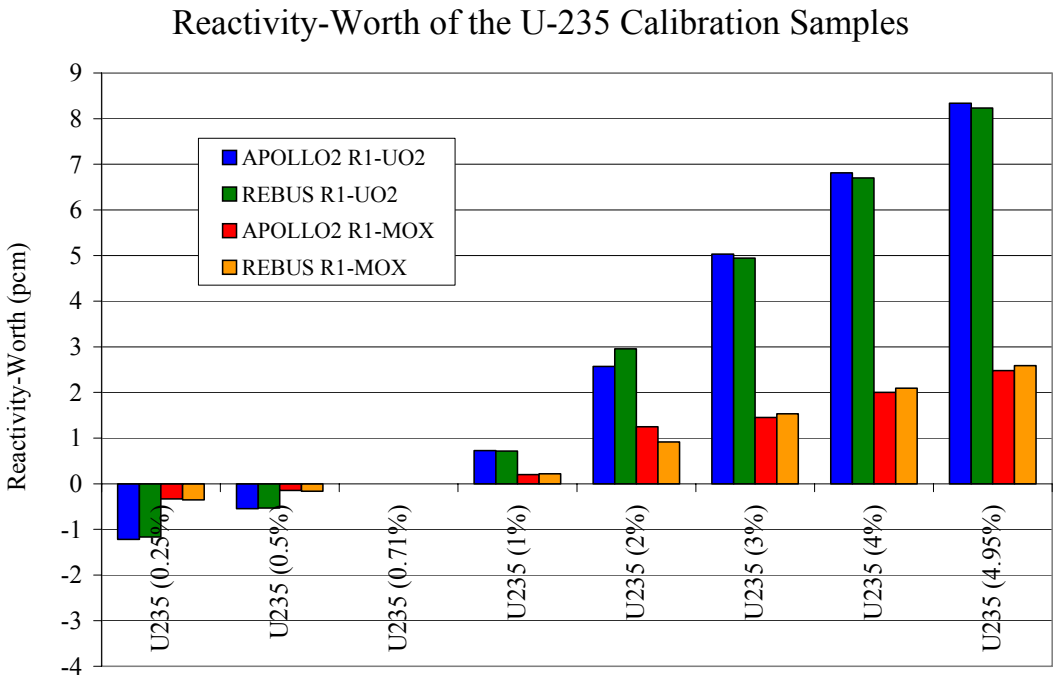

Figure 3

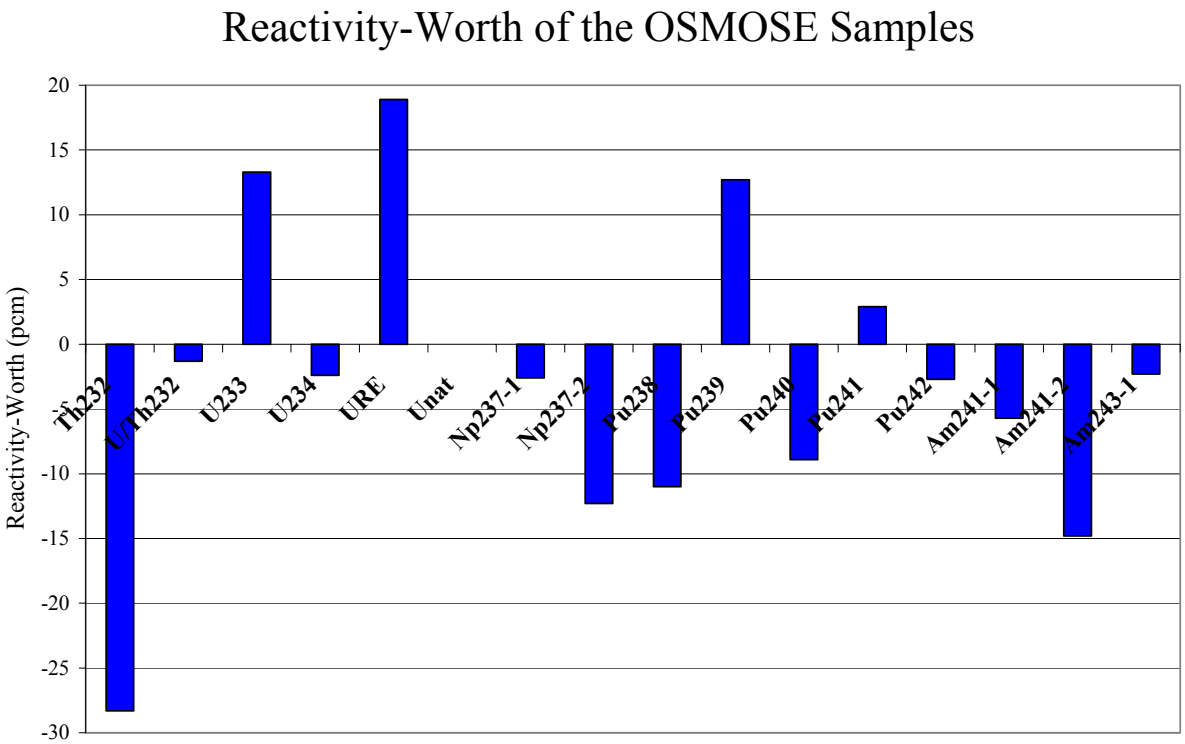

Figure 4

Planned Activities:

Several activities related to reactor modeling are planned for FY06. Pre-analysis estimates of the reactivity effect of the OSMOSE samples in the R1-MOX core configuration will be performed. Additionally, the results of the OSMOSE samples in the R1-UO2 configuration will be studied and re-analyzed if the results of the chemical analysis become available in FY06.

Studies will be performed of the OSMOSE samples in the planned core configurations to determine the relevance of these configurations for the Generation-IV program. Specifically, the configurations will be compared to block-type VHTR and GCR systems.

An evaluation of the OSMOSE samples for use in Doppler-broadening measurements in MINERVE will also be conducted. Results of these efforts will lead to discussions between ANL and CEA on the future direction of the OSMOSE program and other potential related collaborative efforts.

Issues and concerns: None

4. SAMPLE FABRICATION

Objectives:

The OSMOSE program requires the fabrication of 21 oxide samples containing separated actinides (^{232}Th, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, $^{241, 243}$Am and ^{244}Cm, ^{245}Cm). The samples consist of assembled fuel pellets containing the isotopes of interest and a double zircaloy cladding. Specifications for the samples include: the pellet size, the pellet density, the homogeneity of the distribution of the actinides inside the UO2 matrix, and the minimization of contamination during the fabrication process.

Technical Status:

The preparation of the OSMOSE samples is a multi-stage process and includes the purification of the isotope feedstock materials, the creation of an oxide material, the mixing and fabrication of sintered oxide fuel pellets, the assembly of the fuel pellets into the sample, and the welding of the cladding for double-encapsulation.

Isotopes were delivered in 2003 by several suppliers (DOE, CEA, COGEMA and CHEMOTRADE). The isotope supplies required purification to meet the specifications (oxides) and to remove impurities. Table 5 shows the status of the isotope supplies.

All of the isotopes have been purified except the ^{233}U. Because of the high activity due to the presence of decay products of ^{232}U, the operation must be performed in a shielded hot cell. The purification and sample fabrication for the ^{233}U is scheduled at the end of 2006 but may be started sooner in the year. The purification process for the ^{233}U has been established and the fabrication should last 2 or 3 months.

TABLE 5 Status of Isotope Supplies			
Isotope	Quantity (g)	Purification	Supplier
^{232}Th	48 & 2	Not required	CEA
^{233}U	0.5	-	CEA
^{234}U	0.3	Completed	CEA
URT	0.6	Not required	COGEMA
^{238}U	48	Not required	CEA
^{237}Np	0.1 & 0.6	Completed	CEA
^{238}Pu	0.4	Completed	CEA
^{239}Pu	0. 6	Completed	CEA
^{240}Pu	0.15	Not required	DOE
^{241}Pu	0.3	Completed	DOE
^{242}Pu	0,5	Not required	DOE
^{241}Am	0.06 & 0.2	Completed	CEA
^{243}Am	0.1 & 0.5	Not required	DOE
^{244}Cm	2	Not required	CHEMOTRADE
$^{244+245}$Cm	1	Not required	CHEMOTRADE

Pellet Fabrication and Sintering

The oven developed for OSMOSE sample fabrication is running flawlessly and we have achieved 80 thermal cycles without major problems (i.e. 2000 hours in working condition with 300 hours at high temperature).

To prepare for fabrication of the samples in the shielded hot cell, a more efficient tool for the pressing step has been developed. The uniaxial press has been replaced by a new gear based on a tri-part (shell) design (Figure 5). The main component is a common hydraulic press but the special shape of the press chamber allows minimizing the compressive stresses during the opening phase. The densities of the green pellets are also more uniform and the shape is improved. With this new system, it is expected that very few out of specification pellets will be produced. This is critical for fabrication of costly or very active isotopes like curium.

During FY2005, eight complete samples were manufactured including UO_2, UO_2 + $^{232}ThO_2$, UO_2 + $^{234}UO_2$, UO_2 + $^{237}NpO_2$ (1), UO_2 + $^{237}NpO_2$ (2), UO_2 + $^{239}PuO_2$, UO_2 + $^{242}PuO_2$, and $UreO_2$. URE is a sample containing ^{236}U that has been processed from spent nuclear fuel. Table 6 summarizes the characteristics of the first eight samples. With a few minor exceptions, the specifications were obtained for the density and the dimension of the pellet.

Figure 5: New tri-part press with for hot cell with MgO_2 pellet sample

TABLE 6
Metrology of the first set of OSMOSE samples

Sample	Target composition (g)	Mean Density (% of T.D.)	Mean Diameter (mm)	Variance in Diameter (mm)
UO_2		96.1	8.16	0.08
UO_2-^{232}Th	2.0	93.0	8.15	0.09
UO_2-^{234}U	0.3	95.6	8.13	0.08
UO_2-^{237}Np	0.1	95.6	8.15	0.08
UO_2-^{237}Np	0.6	95.1	8.12	0.08
UO_2-^{239}Pu	0.6	93.1	8.15	0.06
UO_2-^{242}Pu	0.5	93.7	8.01	0.20
URE	Pure	96.7	8.10	0.10
Specification		**>95%**	**8.0< Ø <8.2**	**D Ø < 0.1**

TABLE 7 Metrology of sample pellets for second set of OSMOSE samples				
Sample	Target composition (g)	Mean Density (% of T.D.)	Mean Diameter (mm)	Variance in Diameter (mm)
UO_2-^{240}Pu	0.15	97.4	8.10	0.04
UO_2 qualif.[1]	pure	96.0	8.10	0.03
UO_2-^{238}Pu	0.4	92.0	8.16	0.09
UO_2-^{241}Am	0.06	94.7	8.14	0.11
UO_2-^{241}Am	0.2	preparation in progress		
UO_2-^{240}Pu	0.15	scheduled November		
ThO_2	pure	scheduled December		

[1]For each sintering phase, an extra UO2 pellet is placed in the oven for monitoring the quality of the thermal cycle. Systematically the theoretical density was found above 95%.

Pellets for the next set of OSMOSE samples are also currently being fabricated. Table 7 shows the actual status of the preparation and the initial results of the metrology. Fabrication of the second set of samples is expected in the first quarter of FY06, followed by cladding and welding in the second quarter of FY06.

Cladding and Welding

In 2004, all the equipment needed for the cladding and the welding of the samples was installed. Unfortunately, because of technical issues with the laser and the use of the facilities by other programs the qualification of the welding process was not completed until May 2005. After new tests, the system was qualified by the Institut de Soudure.

The cladding and the welding were performed in a very short period of time (8 hours). The sequence was: one reference sample - 4 OSMOSE samples - one reference sample - 4 OSMOSE samples - one reference sample. Figures 6-9 show the cladding and welding process performed in the glove boxes.

After each welding (inner & outer clad) a leak test was performed. Metallographic inspection was performed on the reference samples. The first eight samples were accepted by CEA-Cadarache in June, 2005.

Six samples were delivered to CEA-Cadarache in July, 2005 using a CROFT container. The two remaining samples (Np) were delivered in September, 2005.

Figure 6: Samples before the cladding

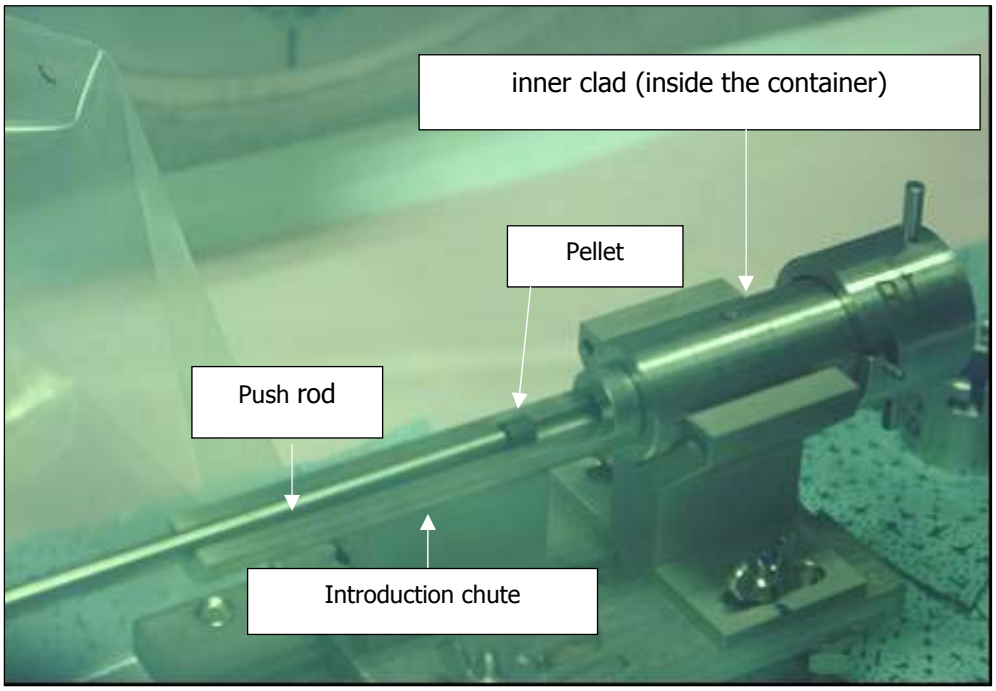

Figure 7: Pellets being inserted into first cladding

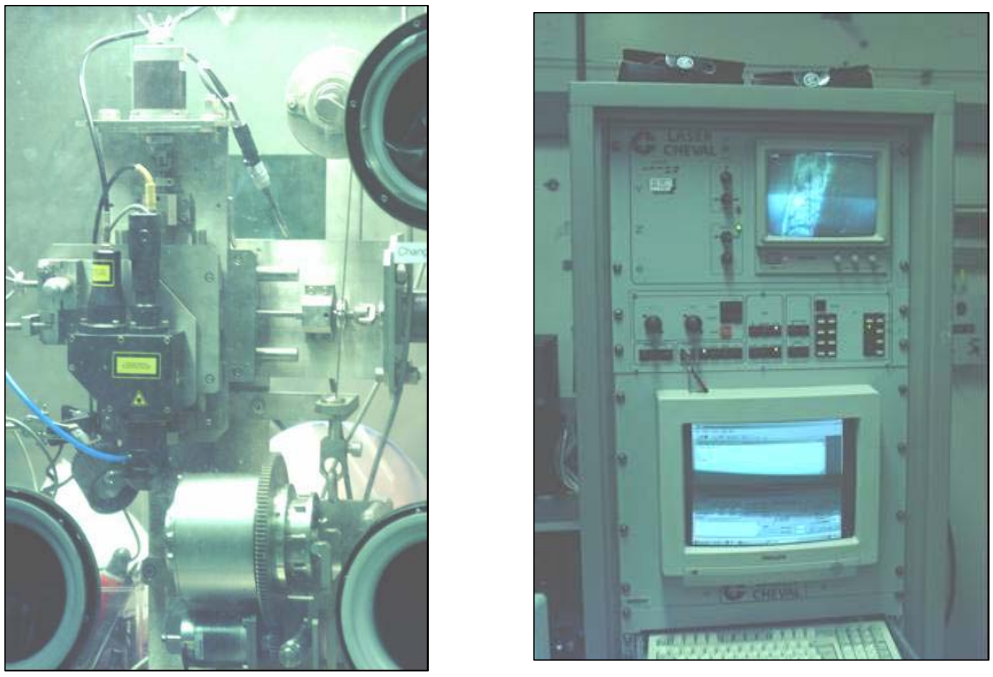

Figure 8: The laser welding station in glove box its remote control

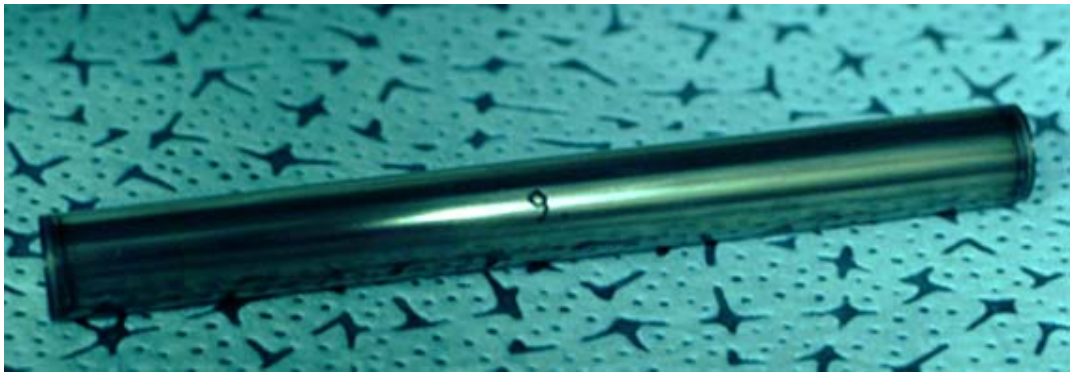

Figure 9: Final sample after welding

Chemical Analysis

The isotopic and chemical analysis of the actinide stocks has been carried out for the remaining actinides. At the present time only ^{243}Am, ^{233}U and curium are still waiting to be analyzed by mass spectrometry techniques (IDMS).

The ^{241}Am level (<500 ppm) has also been checked by alpha spectroscopy in all of the samples.

The analysis of pellets from the first 8 samples began in FY05. The analysis requires the accurate dissolution of the solid sample. Preliminary tests have shown that HNO_3 acid with a small amount of HF acid give excellent results. All the operations are performed by weighing the samples.

The mass of the separated isotope included inside each sample will be checked with a target total uncertainty of 2% by IDMS. Afterward, the concentration of the UO_2 matrix will be determined by a "titriphotogravimetry" method using cerium (IV) titration.

This very accurate method (± 0.1%) is mainly used to analyze the reference samples for quality control of the CEA analytical facilities. The initial data appears close to the expected values.

Planned Activities for FY06:

Finalization of the technical documents regarding the sample fabrication for the first 8 samples will be completed in FY06. This includes the analytical results of the as-fabricated samples and confirmation from analysis at ANL. Reference pellets will be shipped from CEA-Valrho to ANL and then destructively analyzed to confirm the results of analysis performed in France.

The next 6 samples – Pu-240, Pu-238, Am-241 (x2), Pu-241 and Th-232 – will be fabricated in CEA-Valrho and shipped to CEA-Cadarache inside the RD15IIB container in February 2006.

The last 5 samples – Cm-244, Cm-244+245, Am-243 (x 2) and U-233 – will be fabricated and shipped inside the RD15IIB container in 2007. The shipment of these OSMOSE samples from CEA-Valrho to CEA-Cadarache is a major issue because of the high activity of the Curium samples. Thus, CEA-Cadarache has to obtain special authorization to ship the OSMOSE samples. This authorization is expected in November.

Issues and Concerns: None

5. EXPERIMENTS

Objectives:

The main objective of the OSMOSE measurement program is to provide an experimental database of reactivity-worth measurements in different neutron spectra for the heavy nuclides. This database can then be used as a benchmark for integral reactivity-worth measurements to verify and validate reactor analysis codes. Measurements on other samples will provide additional data for benchmarking depletion codes as well as reactor analysis codes (French codes and U.S. codes).

Technical Status:

The OSMOSE measurement program in the MINERVE facility began in July 2005 as scheduled. Before the beginning of the experimental program, several compulsory actions had to be performed including renewal of the safety calculation scheme of the MINERVE facility, presentation of the safety report for the program to the French safety authorities, authorization to begin the program by the French safety authorities, calibration of the

pilot rod, and shipment of the samples from CEA-Valrho to CEA-Cadarache and related authorizations. The final authorizations from the French authorities were provided on June 21, 2005.

Oscillation measurements of the U-235 and borated calibration samples were conducted in July and August. Additional borated calibration samples are being fabricated and will be oscillated in October, 2005. Oscillation measurements using the first 8 OSMOSE samples began in September, 2005. Technical status of each step in the process is detailed below.

Calibration of the pilot rod

The ability of the oscillation technique to accurately determine the reactivity-worth of unknown samples relies on an accurate calibration and understanding of the reactivity effects from the operation of the pilot rod. The pilot rod is a servo-driven system that rotates cadmium sections in overlapping patterns to cause a change in the neutron absorption of the pilot rod as a function of the angle of the rotor. Because of the overlapping cadmium regions and the rotation of the cadmium sections, the effect on reactivity is not proportional to the rotor position for all angles of rotation. The calibration of the pilot rod is necessary to determine the range of angles of rotation of the rotor that are proportional to reactivity, and to accurately determine the differential change in reactivity. The technique does not determine the absolute value of reactivity for a given rotor position, but instead is based on the relative reactivity effect, which is significantly more accurate for determining small changes in reactivity.

To calibrate the pilot rod for oscillation measurements, two stages of calibration were performed. The first stage dealt with verifying that the reactivity range of the pilot rod matches the range of the sample reactivity, i.e. ± 0.0001 (10 pcm). This was accomplished by positioning the pilot rod at different angles (i.e. different values of voltage on the rotor) and measuring the reactivity excess of the core. By doing this over the entire range of angles, a calibration curve of the pilot rod is created, as shown in Figure 10. This is a crude calibration that is adequate for initial positioning of the pilot rod but not sufficient for detailed measurements of small reactivity changes.

In the second stage of the calibration, the differential reactivity effect was determined for small changes in the voltage applied to the pilot rod. The position of the pilot rod was controlled by the bias voltage applied to the rotor. As observed in Figure 10, the change in reactivity effect is not directly proportional to the voltage. This means that the differential effects of applying a constant voltage to the rotor is also not a constant effect, i.e. it depends on the initial positioning of the rotor (or voltage applied to the rotor). This initial voltage is equivalent to the mean amplitude during the oscillations of the samples. So a calibration curve was created which relates the variation of the angle of the pilot rod (i.e. amplitude of the signal) to its mean angle (mean value of the signal). This relation is linear over a small range and allows the normalization of all measurements to a specified reference angle. Figure 11 shows the calibration curve of the pilot rod in the R1-UO2 configuration.

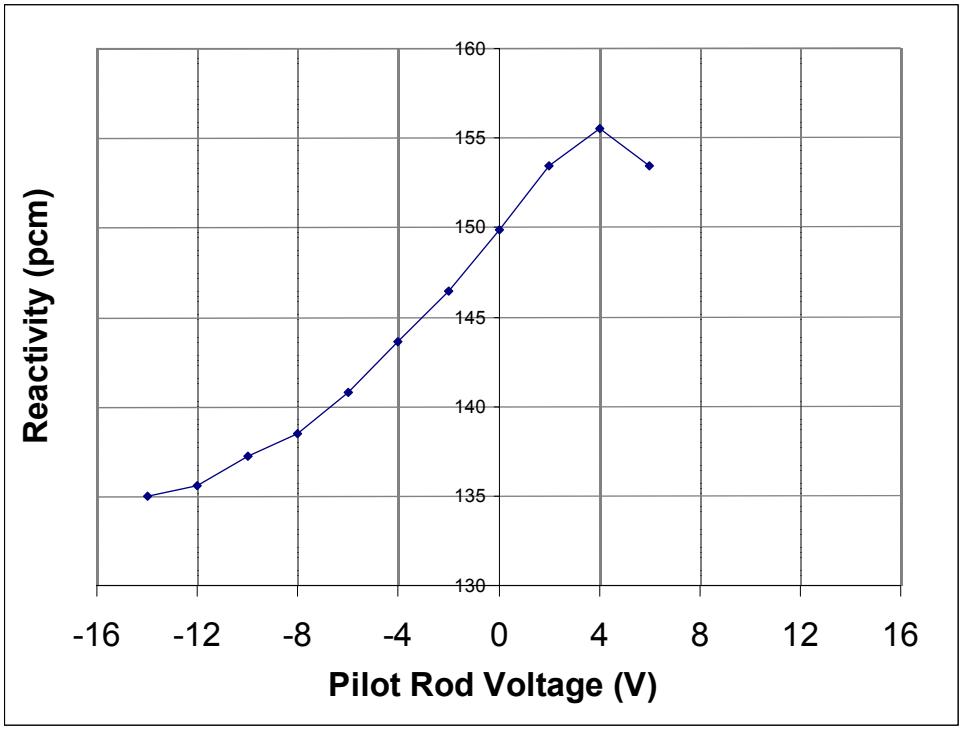

Figure 10: Reactivity of the pilot rod in the R1-UO2 configuration of the OSMOSE program

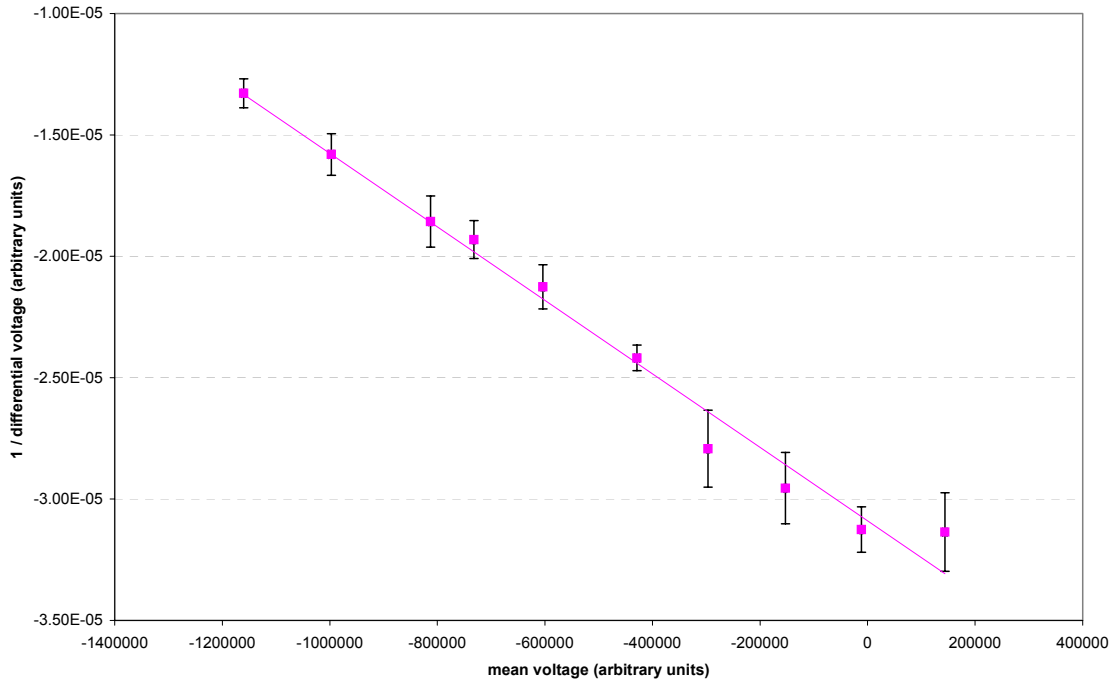

Figure 11: Differential efficiency curve of the pilot rod in the R1-UO2 configuration of the OSMOSE program

In this region of linearity, the response from all samples can be directly compared based on the same reference angle θ_0 using the following relationship:

$$f(\theta) = f(\theta_0) \times (1 + K \times (\theta - \theta_0))$$

with $f(\theta)$ = measured amplitude, θ = mean position of the pilot rod during the measurement, θ_0 = reference mean position (chosen in the middle of the linear part of the differential efficiency curve), $f(\theta_0)$ = amplitude of the signal if the measurement had been performed with a mean position of the pilot rod equal to θ_0, and K = constant dependant on θ_0 and on the linear equation of the differential efficiency curve.

The pilot rod calibration allows the reference angle θ_0 to be established and the normalization factor K to be determined. This calibration then allows all of the oscillation measurements to be normalized to the same reference angle.

Calibration curves for sample measurements

Once the response of the pilot rod was calibrated, the signal was calibrated using reference samples. There are two series of calibration samples, one contains a UO2 matrix with different uranium enrichments (natural, 0.25%, 0.5%, 0.72%, 1%, 2%, 3%, 4% and 4.95% U235) and the other contains a UO2 matrix (0.25% or 0.53% enriched in U-235) with a range of boron concentrations (0, 60, 150, 333, 419, and 1062 ppm).

Figure 12 and Figure 13 show the calibration curves obtained in the R1-UO2 configuration of the OSMOSE program. Table 8 and Table 9 show the raw data for the oscillation of the calibration samples.

The reactivity worth of the calibration samples is obtained by deterministic calculations with an accuracy of approximately 1 %. Thus, the reactivity worth of every sample that is oscillated in MINERVE can be determined by comparing the response to the response from the calibration samples. Samples with a positive reactivity effect are compared to ^{235}U calibration samples, and samples with negative reactivity effects are compared to the boron calibration samples. The total uncertainty on the calibration measurements, involving material balance of the samples, reproducibility of the measurement and uncertainty on the reactivity worth calculation, is about 2 %.

Note that another series of new borated calibration samples, specially fabricated for the OCEAN program (Oscillation in Core of Samples containing New Absorbers) and the OSMOSE programs is already available, and will be oscillated in October in the R1-UO2 configuration. The new borated samples are made of a UO2nat matrix with a range of boron concentrations (100, 200, 300, 400, and 500 ppm). A better knowledge of their material balance should improve the accuracy on the calibration of the signal for the borated samples.

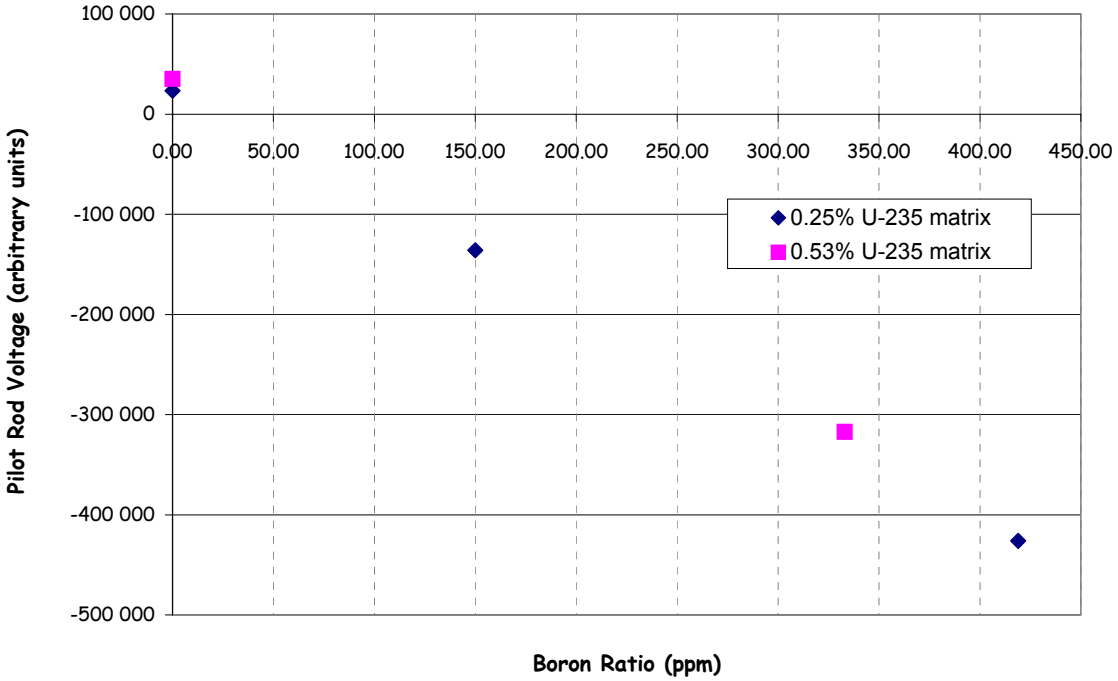

Figure 12: Calibration curve for boron loaded samples in the R1-UO2 configuration of the OSMOSE program

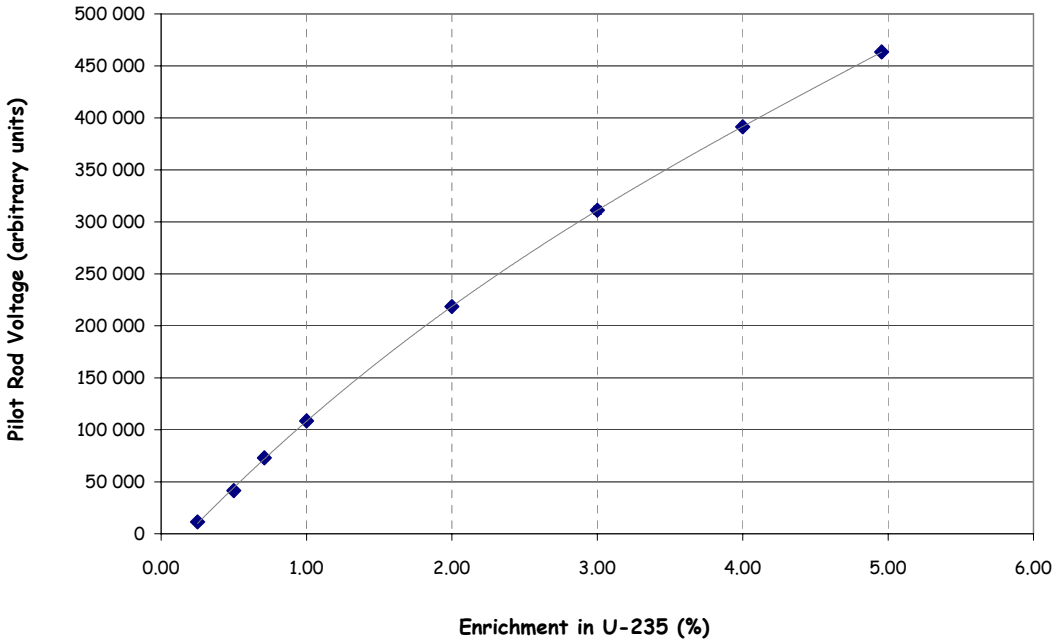

Figure 13: Calibration curve for 235U calibration samples in the R1-UO2 configuration of the OSMOSE program

TABLE 8: Raw data for the borated calibration samples

Name	Measurement Date	Signal (arbitrary Unit)	s.d. (arbitrary Unit)
7 (0 ppm)	28/07/2005	23 314	1 932
7 (0 ppm)	29/07/2005	21 917	2 893
7 (0 ppm)	04/08/2005	22 657	1 755
7 (0 ppm)	05/08/2005	23 368	1 742
7 (0 ppm)	11/08/2005	25 269	1 576
9 (150 ppm)	27/07/2005	-133 270	2 323
9 (150 ppm)	29/07/2005	-134 666	2 082
9 (150 ppm)	03/08/2005	-132 646	2 050
9 (150 ppm)	05/08/2005	-138 260	2 166
9 (150 ppm)	10/08/2005	-136 294	2 425
9 (150 ppm)	12/08/2005	-139 832	1 692
10 (419 ppm)	27/06/2005	-429 307	1 733
10 (419 ppm)	30/06/2005	-426 642	2 054
10 (419 ppm)	20/07/2005	-425 187	1 158
10 (419 ppm)	21/07/2005	-426 829	2 576
10 (419 ppm)	25/07/2005	-425 400	2 188
10 (419 ppm)	12/08/2005	-423 287	1 787
33 (333 ppm)	01/07/2005	-315 814	1 083
33 (333 ppm)	22/07/2005	-316 052	1 815
33 (333 ppm)	19/07/2005	-316 623	3 522
33 (333 ppm)	21/07/2005	-318 730	3 654
33 (333 ppm)	26/07/2005	-318 651	2 659
32 (0 ppm)	30/06/2005	35 151	1 968
32 (0 ppm)	06/07/2005	34 325	1 848
32 (0 ppm)	27/07/2005	37 563	1 929
32 (0 ppm)	28/07/2005	34 655	2 374
32 (0 ppm)	20/07/2005	35 505	2 916
32 (0 ppm)	22/07/2005	34 007	3 116

TABLE 9: Raw data for the U-235 calibration samples

Name	Measurement Date	Signal (arbitrary Unit)	s.d. (arbitrary Unit)
H1 (0.25% U-235)	29/06/2005	10 910	2 002
H1 (0.25% U-235)	07/01/2005	12 046	1 774
H1 (0.25% U-235)	21/07/2005	11 260	1 942
H1 (0.25% U-235)	26/07/2005	11 021	2 557
H1 (0.25% U-235)	12/09/2005	8 705	3 095
H3 (0.71% U-235)	30/06/2005	70 583	1 139
H3 (0.71% U-235)	04/06/2005	73 666	2 053
H3 (0.71% U-235)	19/07/2005	74 867	1 344
H3 (0.71% U-235)	26/07/2005	71 796	2 120
H3 (0.71% U-235)	23/08/2005	74 445	1 863
H3 (0.71% U-235)	13/09/2005	71 757	2 003
H5 (2% U-235)	27/06/2005	220 579	1 844
H5 (2% U-235)	01/07/2005	215 853	2 381
H5 (2% U-235)	21/07/2005	218 901	1 561
H5 (2% U-235)	25/07/2005	216 855	1 915
H5 (2% U-235)	20/07/2005	221 448	2 257
H5 (2% U-235)	23/08/2005	217 912	1 792
H8 (4.955% U-235)	21/07/2005	465 982	2 224
H8 (4.955% U-235)	27/06/2005	462 406	3 661
H8 (4.955% U-235)	22/07/2005	463 511	3 078
H8 (4.955% U-235)	27/07/2005	461 220	3 814
H8 (4.955% U-235)	23/08/2005	463 498	2 099
H2 (0.50% U-235)	29/07/2005	41 400	2 277
H2 (0.50% U-235)	27/07/2005	43 032	2 392
H2 (0.50% U-235)	03/08/2005	42 189	1 488
H2 (0.50% U-235)	05/08/2005	40 983	1 498
H2 (0.50% U-235)	10/08/2005	40 130	1 707
H4 (1% U-235)	28/07/2005	107 563	2 018
H4 (1% U-235)	04/08/2005	108 984	1 649
H4 (1% U-235)	05/08/2005	110 913	1 853
H4 (1% U-235)	10/08/2005	108 062	2 220
H4 (1% U-235)	11/08/2005	107 194	1 424
H6 (3% U-235)	28/07/2005	315 427	2 246
H6 (3% U-235)	02/08/2005	307 077	1 617
H6 (3% U-235)	04/08/2005	311 341	2 462
H6 (3% U-235)	10/08/2005	314 276	2 337
H6 (3% U-235)	11/08/2005	308 138	2 849
H7 (4% U-235)	02/08/2005	392 303	1 532
H7 (4% U-235)	05/08/2005	388 716	2 448
H7 (4% U-235)	10/08/2005	389 641	1 614
H7 (4% U-235)	11/08/2005	394 818	1 914
H7 (4% U-235)	23/08/2005	391 526	3 359

Oscillation of the OSMOSE samples

The oscillation of the first series of the OSMOSE samples began on September 1, 2005. Each of the 8 samples was oscillated 5 or 6 times inside the R1-UO2 configuration during September 2005. A measurement consists of 10 cycles with a cycle length of 120 seconds. Table 10 shows the preliminary results of the first oscillations. Figure 14 shows an example of the signal obtained by oscillating the Pu-239 sample.

Planned Activities:

The OSMOSE program is performed in the R1-UO2 core configuration in parallel to the OCEAN (Oscillation in Core of samplEs containing New Absorbers) program (funded by EDF and CEA).

The oscillation of the first 8 OSMOSE samples will be finished in October, 2005. The oscillation of the second set of OSMOSE samples (Pu-240, Pu-238, Am-241 (x2), Pu-241 and Th-232) – will be performed in February 2006, and should be finished by April 2006.

The core will be reloaded with the R1-MOX configuration and measurements using the first two sets of OSMOSE samples plus the remaining samples will be performed from July 2006 through March 2007. Oscillation of the last 5 OSMOSE samples (Am-243 (x2), Cm-244, Cm-244+245 and U-233) in the R1-UO2 configuration is scheduled from April, 2007 through June, 2007.

The OSMOSE samples will be oscillated in the R2-UO2 configuration from July, 2007 to March, 2008 and in the MORGANE-R configuration from April, 2008 to December, 2008.

Issues and concerns: None

Table 10: Preliminary raw data for the OSMOSE samples

Name	Measurement Date	Signal (arbitrary Unit)	s.d. (arbitrary Unit)
U/Th-232	02/09/2005	18 015	1 680
U/Th-232	08/09/2005	20 718	2 551
U/Th-232	13/09/2005	18 580	1 715
Pu-239	02/09/2005	249 491	2 584
Pu-239	08/09/2005	248 998	2 750
Unat	02/09/2005	68 557	2 556
Unat	08/09/2005	56 882	1 077
Unat	13/09/2005	62 146	1 657
URE	07/09/2005	350 439	2 762
URE	19/09/2005	360 563	1 851
Pu-242	06/09/2005	-2 225	2 045
Pu-242	08/09/2005	-3 308	2 631
U-234	06/09/2005	-6 910	1 864
U-234	12/09/2005	-5 885	2 115

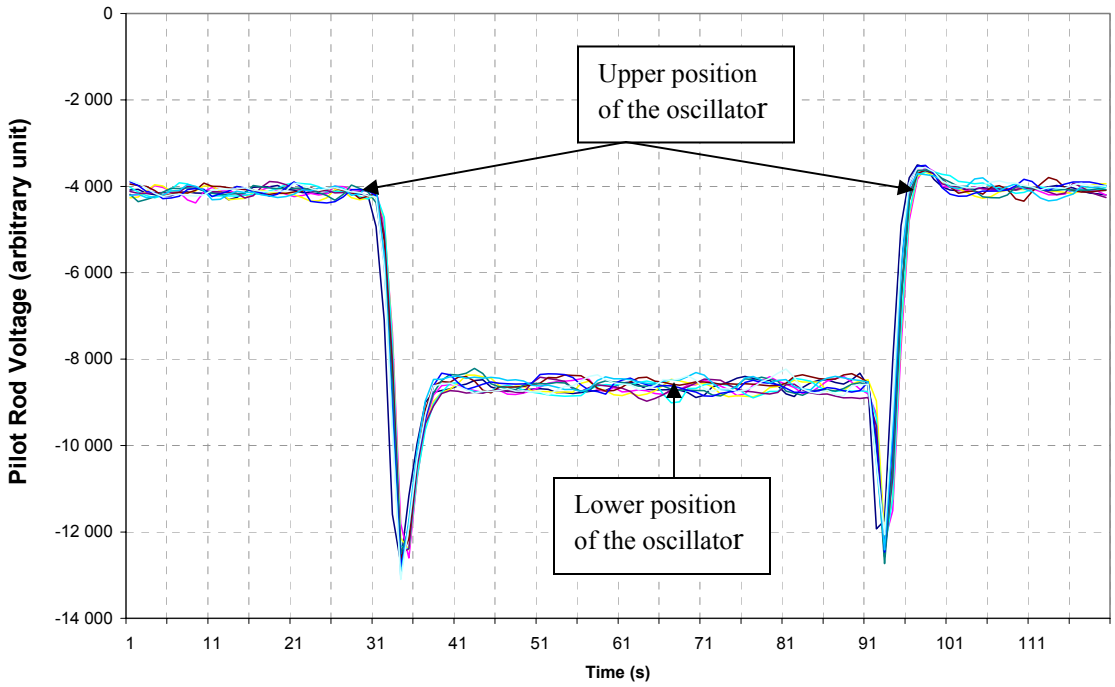

Figure 14: Signal of the Pu-239 OSMOSE sample during its oscillation in MINERVE R1-UO2 Configuration

6. DATA ANALYSIS

Objectives:

The data analysis tasks address the analysis and reduction of data for each series of measurements for the different core configurations. In general, the data analysis tasks include the review and analysis of the raw data for the full range of separated samples for the OSMOSE program in each reactor configuration, the analysis of the raw data from the calibration and test measurements performed with calibration samples of differing uranium and boron compositions, and the analysis of the raw data for all spectral indices and axial and radial distributions measurements performed to support the OSMOSE program.

In addition, the data analysis tasks include processing the raw data from the instruments and data acquisition system to produce the experimental results, i.e. the analysis and reported sample-worth for each of the separated samples and calibration samples for all reactor configurations included in the OSMOSE program.

The objective of the data analysis tasks are to compile the data and results into a reactor physics benchmark (in accordance with guidance provided by the American Nuclear Society Joint Benchmark Committee), This benchmark includes a thorough review and study of the systematic errors in the measurement technique, measurements, and statistical uncertainties. In addition, cross comparisons between US and French calculational results will be performed.

Technical Status:

Data analysis activities related to pre-planning of the measurements and to support the calibration of the pilot rod and sample calibrations was discussed in previous sections.

Treatment of the data from the oscillation measurements of the first set of OSMOSE samples has not begun.

Planned Activities:

Significant data analysis activities are planned for FY06. The measurement data for the first set of OSMOSE samples will be completely treated and a first benchmark report will be drafted. The issuance of the report may be delayed pending the chemical analysis and confirmation of the sample compositions.

In addition, estimates of the reactivity effect of the samples in the R1-MOX configuration will be performed prior to the oscillation measurements in the R1-MOX loading.

Analyses will be performed to address several key technical questions. Specifically, the relevance of the OSMOSE configurations to the block-type VHTR, the GCR, and other

Gen-IV concepts will be studied. The neutron spectra of the different OSMOSE configurations will be compared to the representative spectra for the Gen-IV concepts.

An evaluation of the use of the OSMOSE samples in the MINERVE reactor for Doppler-broadening measurements will also be conducted.

Issues and Concerns: None

7. SUMMARY AND CONCLUSIONS

The goal of the OSMOSE program is to measure the reactivity effect of minor actinides in known neutron spectra of interest to the Generation-IV reactor program and other programs and to create a database of these results for use as an international benchmark for the minor actinides. The results are then compared to calculational models to verify and validate integral cross-sections for the minor actinides.

The OSMOSE program includes all aspects of the experimental program – including the fabrication of fuel pellets and samples, the oscillation of the samples in the MINERVE reactor for the measurement of the reactivity effect, reactor physics modeling of the MINERVE reactor, and the data analysis and interpretation of the experimental results.

Significant accomplishments in FY05 include: **1**. The fabrication of eight of the OSMOSE samples at CEA-Valrho and shipment of the 8 samples to CEA-Cadarache. Six samples were shipped in July and two Np samples were shipped in September**. 2.** Calculations and experiments in support of characterization of the MINERVE reactor and for safety authorization were completed. Calibration of the pilot rod and measurements using the U-235 and Boron calibration samples were performed in the R1-UO2 configuration. **3.** An initial series of calculations of the reactivity-worth of the samples in the R1-UO2 core configuration were completed using the REBUS model. The calculations are based on the specifications for fabrication, so they are preliminary until sampling and analysis have been completed on the fabricated samples. The estimates indicate a range of reactivity effect from -22 pcm to +25 pcm compared to the natural U sample. **4. Oscillation measurements using the first 8 OSMOSE samples began in September in the R1-UO2 configuration.**

8. FUTURE ACTIVITIES AND CONTINUED COLLABORATION

The continuation of the DOE/CEA collaboration on the OSMOSE program includes the participation of DOE in the conduct of the experiments and the development and comparison of analytic tools and models of CEA and DOE based on Monte Carlo and deterministic methods. CEA continues to support and fund the experimental and analytical programs at the CEA Cadarache Research Center. The U.S. involvement in the program is being supported in 2006 as part of the ANL-Model Improvement work package within the Generation-IV program. The project is also continuing as a CEA/DOE collaboration within the guidelines of the new I-NERI program.

Safety authorization will be requested for the R2-UO2 and the MORGANE-R core configurations. In particular, the adaptation of the new calculation scheme to the MORGANE-R loading (MOX 11% fuel pins in an hexagonal pitch) will have to be performed. Furthermore, a separate safety authorization will be needed for measuring the Curium samples in all the core configurations.

Within the framework of the GEN-IV future concepts and of the OSMOSE I-NERI collaboration, ideas for extending the collaboration have been evoked during 2005. There are several critical issues that need to be addressed within the framework of the OSMOSE program, the Gen-IV program, and future collaborations with CEA. Specifically, the questions that need to be addressed are:

1. How do the spectra in the MINERVE core loadings compare to representative spectra of the Gen-IV reactor concepts.
2. If the spectra do not compare, what would be the approach for developing a new core configuration that would allow the spectra to closely match the spectra for the Gen-IV concepts?
3. For harder neutron spectra (like in the MORGANE-S and ERMINE loadings or in new loadings), what types of samples are necessary for accurate measurements?
4. What would be the approach for fabricating additional samples for these configurations?
5. Is there a programmatic interest in conducting Doppler-broadening measurements?
6. Can the OSMOSE samples and MINERVE facility be used to perform Doppler-broadening measurements? And if so, what types of modifications would be required?

A working group between CEA and ANL will be created in FY06 to study these issues and recommend a path forward to address the feasibility of such program extensions. The working group will issue a report by the end of FY06.

9. MILESTONES

Milestones for the OSMOSE program are defined in work package number G-A0802K01. The milestones and deliverables are excerpted from the work package and listed below. All milestones and deliverables have been met in FY05.

Work Package No: G-A0802K01

Schedule/Budget Info:

Act. No.	A/M/D*	Description	Start Date	Fin. Date
10	A	Reactor modeling and reactivity-worth estimates of OSMOSE samples in R1-UO2 configuration	10/1/2004	6/30/2005
11	M3	Issue report on reactivity-worth estimates of OSMOSE samples in R1-UO2 configuration		6/30/2005
12	D	Report on reactivity-worth estimates in R1-UO2 configuration		6/30/2005
13	A	Collaborate with CEA personnel and monitor progress of sample fabrication activities	10/1/2004	9/30/2005
14	A	Support experimental measurements in R1-UO2 configuration	3/1/2005	9/30/2005
15	M3	Issue annual report of measurement activities in R1-UO2		9/30/2005
16	D	Status report on OSMOSE measurements in R1-UO2 configuration		9/30/2005

* A - Activity, M - Milestone, D - Deliverable

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